Kinetics of Nuclear Decay

Nuclear Decay Types

There are many unstable nuclei - in nature

Nuclear Science began with Henri Becquerel’s discovery (1896) of uranium radioactivity

and man-made:

$$^{30}Al(\alpha,n)^{30}P \xrightarrow{\beta^+} {}^{30}Si \quad \text{Joliot \& Curie}, 1934$$

$$Th-\alpha \text{ source}, E_\alpha \approx 6\,MeV$$

$$^A_Z X_N$$

N neutron number
Z proton number
A=N+Z

Types of decay:

$$\alpha \text{ decay} \quad : \quad {}^A_Z X_N \to {}^{A-4}_{Z-2}Y_{N-2} + \alpha$$

$$\beta^- \text{ decay} \quad : \quad {}^A_Z X_N \to {}^A_{Z+1}Y_{N-1} + e^- + \bar{\nu}_e$$

$$\beta^+ \text{ decay} \quad : \quad {}^A_Z X_N \to {}^A_{Z-1}Y_{N+1} + e^+ + \nu_e$$

$$e^- \text{ capture} : \quad {}^A_Z X_N + e^- \to {}^A_{Z-1}Y_{N+1} + \nu_e$$

$$\mu^- \text{ capture} : \quad {}^A_Z X_N + \mu^- \to {}^A_{Z-1}Y_{N+1} + \nu_\mu$$

(β⁻ decay, β⁺ decay, e⁻ capture, μ⁻ capture:) “weak” interactions

$$\gamma \text{ decay} \quad : \quad {}^A_Z X^{**}_N \to {}^A_Z X^{*}_{NN} + \gamma$$

$$\text{Fission} \quad : \quad {}^A_Z X_N \to {}^{A_1}_{Z_1}F_{N_1} + {}^{A-A_1-x-y}_{Z-Z_1-y}F_{N-N_1-x} + xn + yp$$

Various rare heavy particle (cluster) decays

Beta Decays of Odd-A and Even-A Nuclei

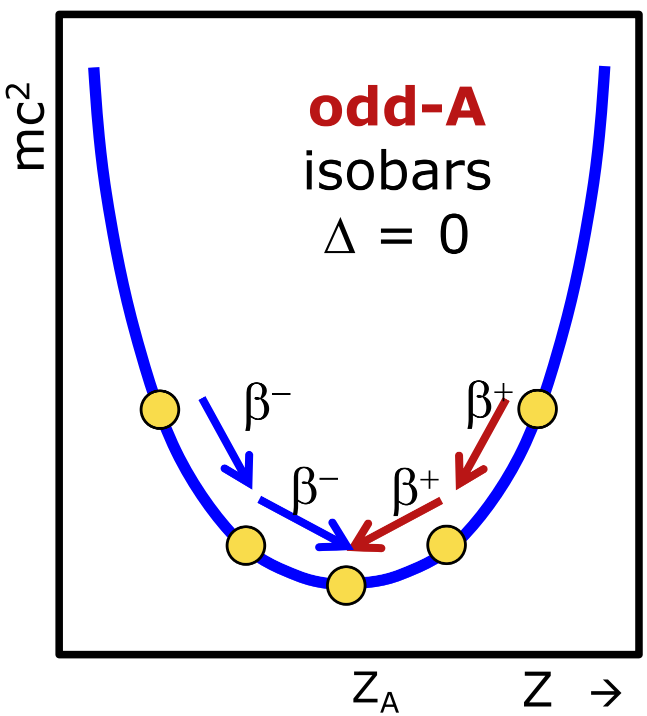
$$m(A,Z) = \alpha(A) - \beta(A)Z + \gamma(A)Z^2 \pm \Delta$$

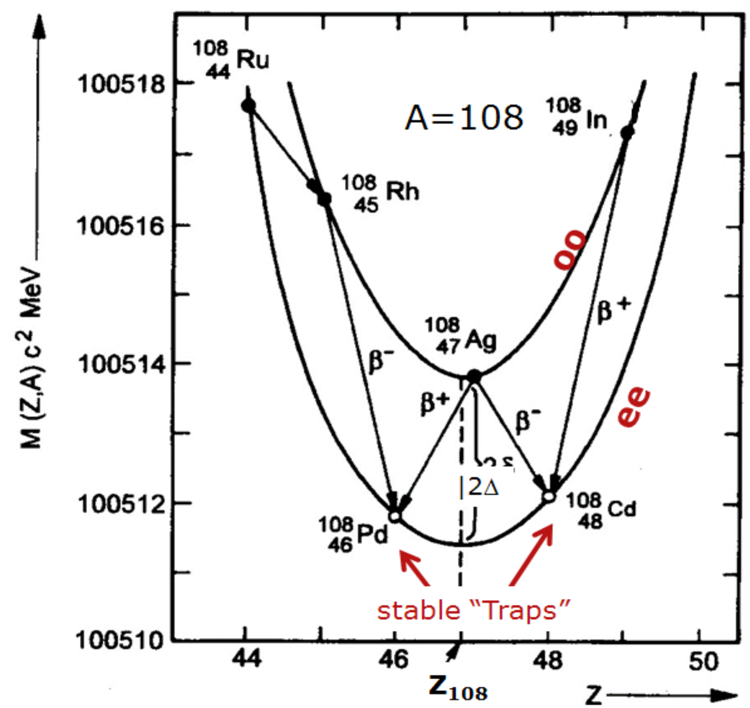
$$m = m_{min} : Z_A = \frac{\beta}{2\gamma} = \frac{\left[4a_s + \left(m_n - m_p - m_e\right)c^2\right]A}{2\left(4a_s + a_C A^{2/3}\right)}$$

$$\Delta = \begin{cases} +\frac{11.2}{\sqrt{A}} MeV & o-o \\ 0 \quad MeV & A = odd \\ -\frac{11.2}{\sqrt{A}} MeV & e-e \end{cases}$$

Expand around Z_A:
Mass parabola bottom of valley

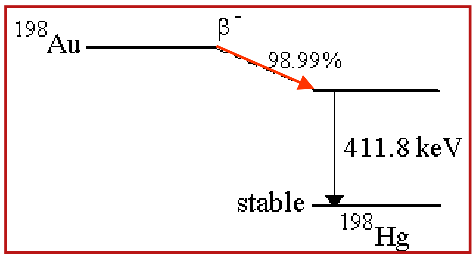
$$m(Z) \approx \left[\tilde{\alpha}(A) \pm \Delta\right] + \tilde{\beta}\left(Z - Z_A\right)^2$$





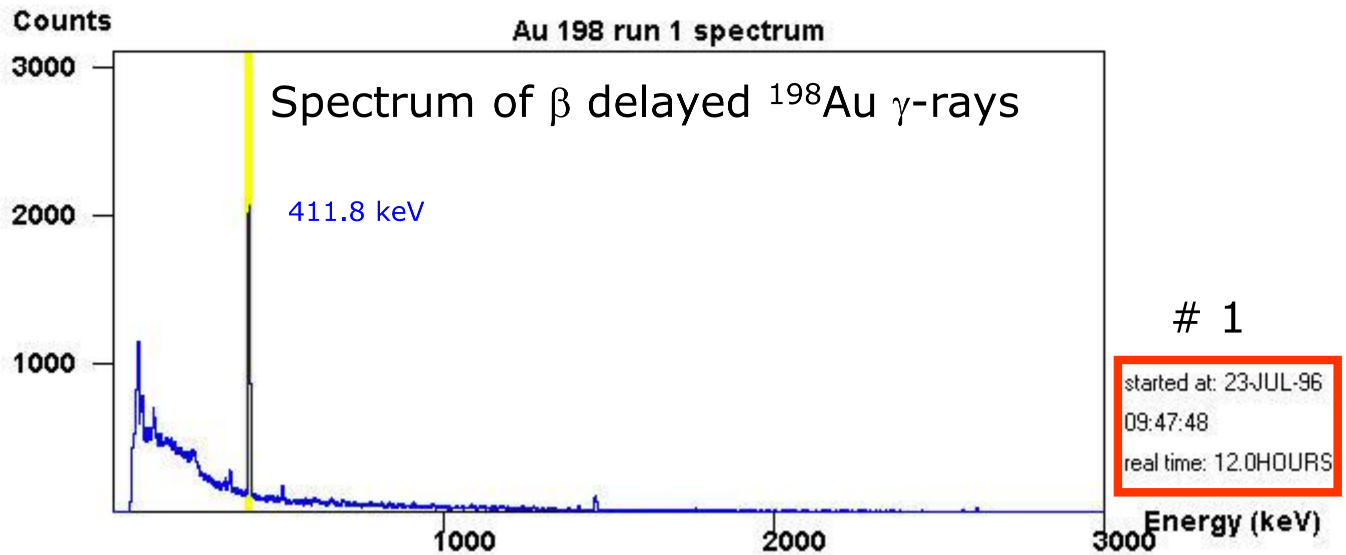
Observing a Finite Lifetime of the ^{198}Au g.s.

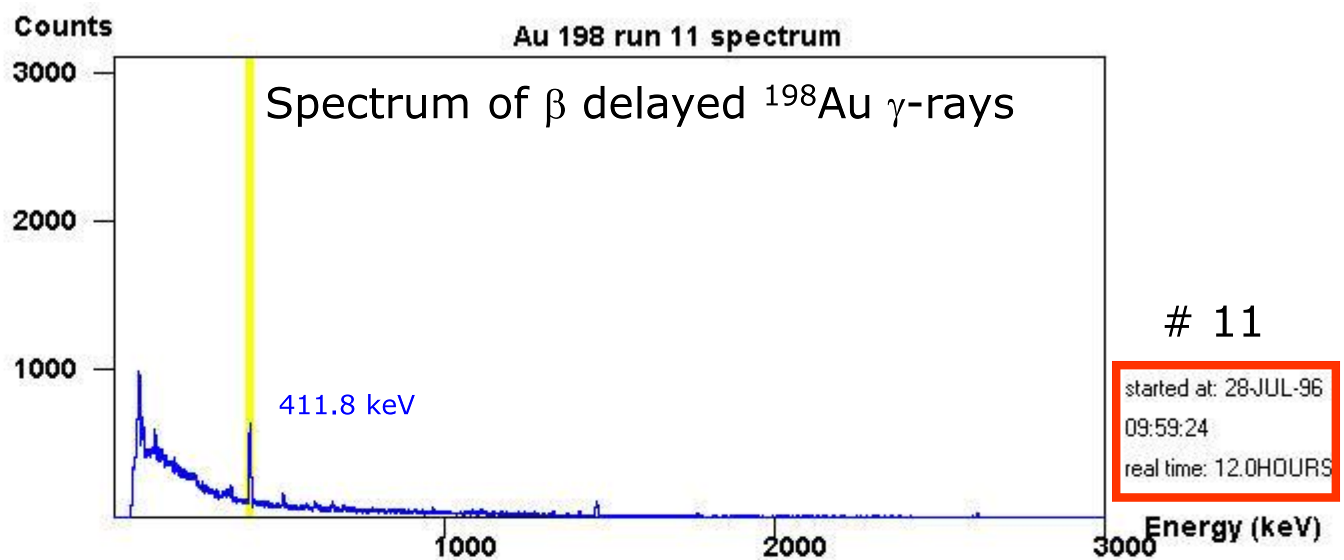
E. Norman et al., http://ie.lbl.gov/radioactive decays/page2



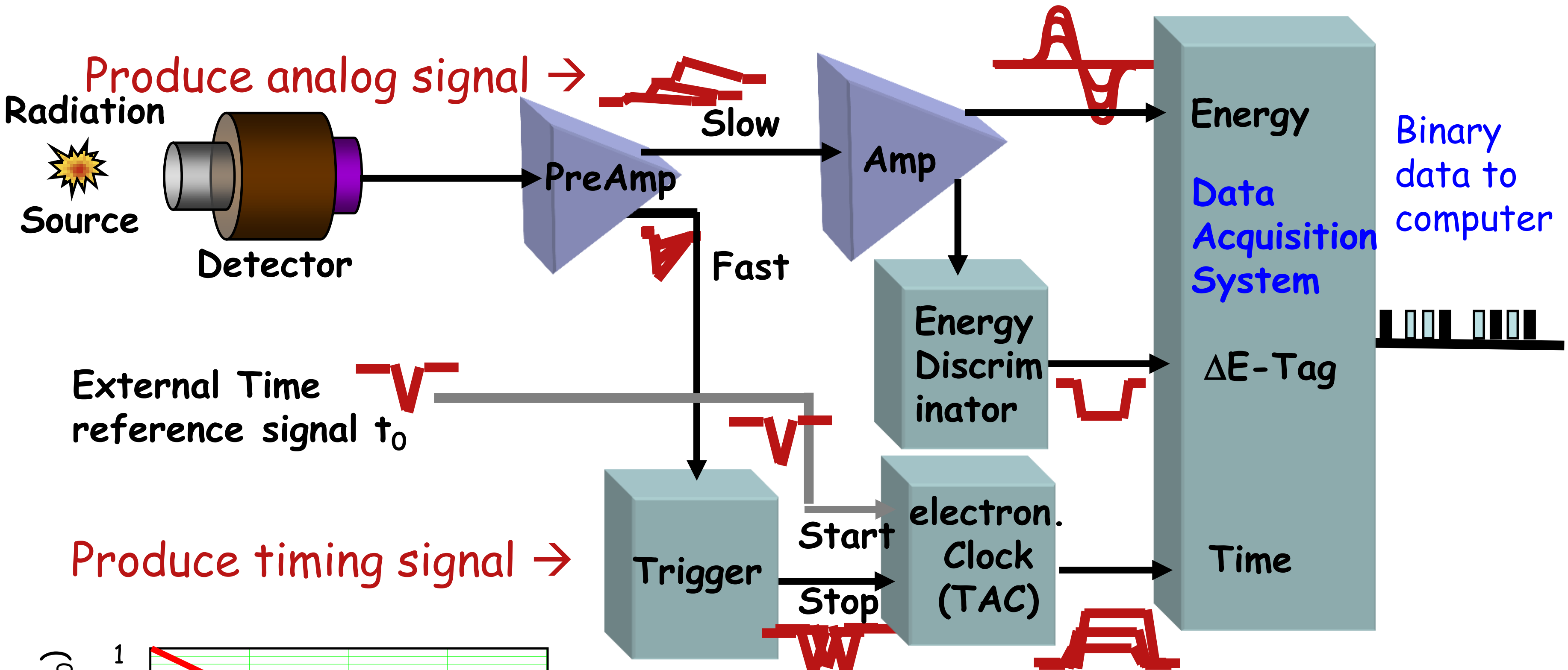
γ decay of ^{198}Hg exc. state is prompt: $\tau_\gamma \ll \tau_\beta$

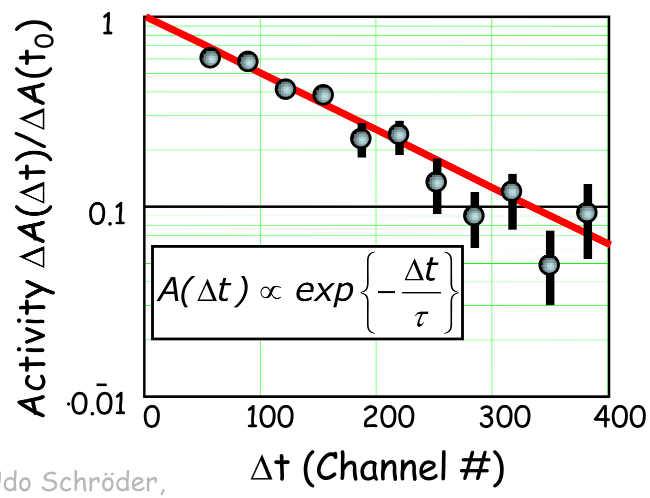
11 measurements
Each spectrum ran for 12 hours real time
#11 taken
5 days after #1





Measuring "Decay Curves": Fast-Slow Signal Processing



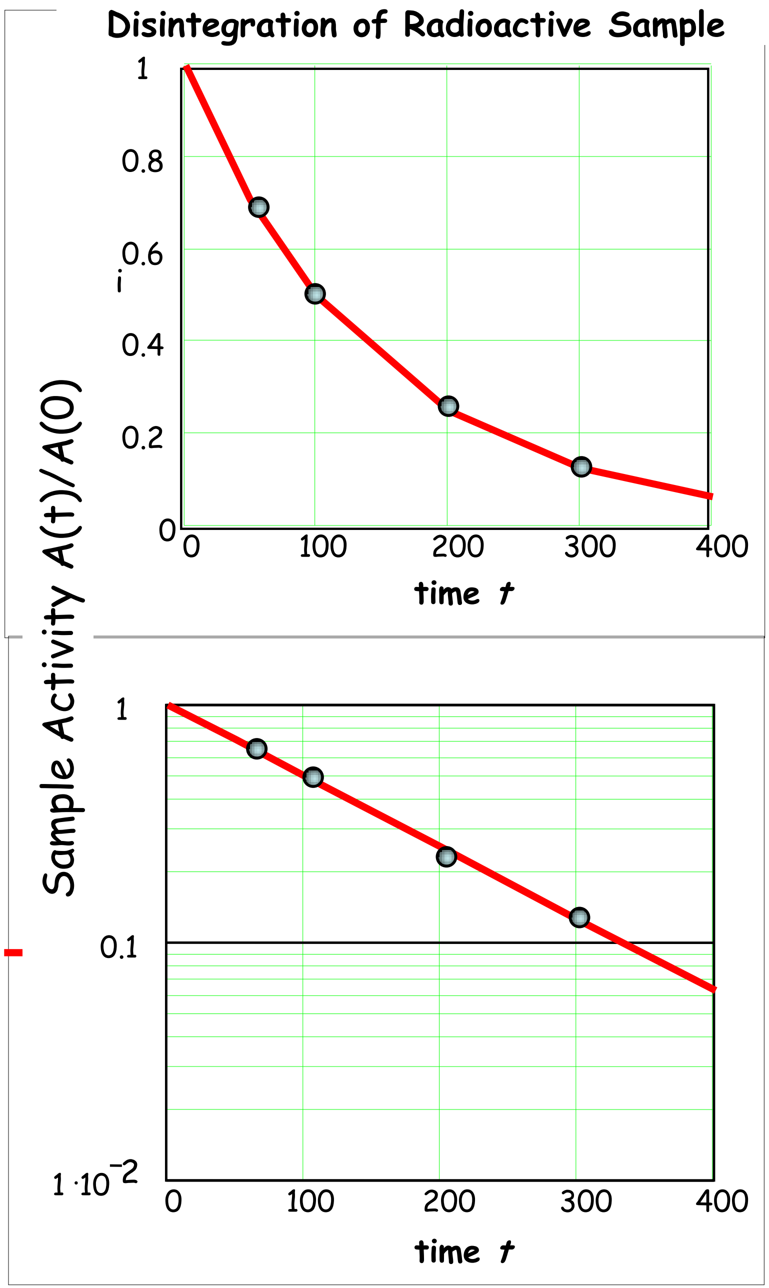


Measured: Energy and time of arrival $\Delta t = t - t_0$ (relative to an external time-zero t_0) for radiation (e.g., γ-rays), energy discriminator to identify events (ΔA) in a certain energy interval ΔE by setting an identifier "tag."

Calibrate Δt axis channel # → time units (s, y,..)

Watch that Δt-channel ≪ τ.

Kinetics of Nuclear Decay: Logarithmic Decay Law



First – order process:

$$\text{Activity} = \#\text{of decays / unit time}$$

$$A = -\dot{N} = -\frac{d}{dt}N(t) = \lambda \cdot N$$

exponential law (*base* $e = 2.1828..$)

$$N(t) = N(t=0)\cdot e^{-\lambda \cdot t} \quad \text{life time } \tau = \frac{1}{\lambda}$$

exponential law (*base* 10)

$$N(t) = N(t=0)\cdot 10^{-\frac{\lambda}{2.303}\cdot t}$$

exponential law (*base* 2)

$$N(t) = N(t=0)\cdot 2^{-\frac{\lambda}{0.6931}\cdot t}$$

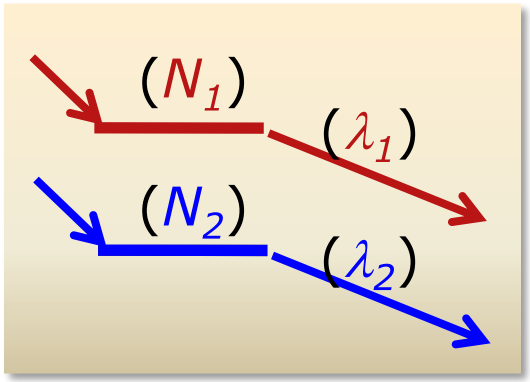
$$\text{Half life } t_{1/2} = \frac{0.6931}{\lambda}$$

$$\text{Decay width } \Gamma := \frac{\hbar}{\tau} = \hbar \cdot \lambda$$

Sum Radioactivity

Genetically independent species:

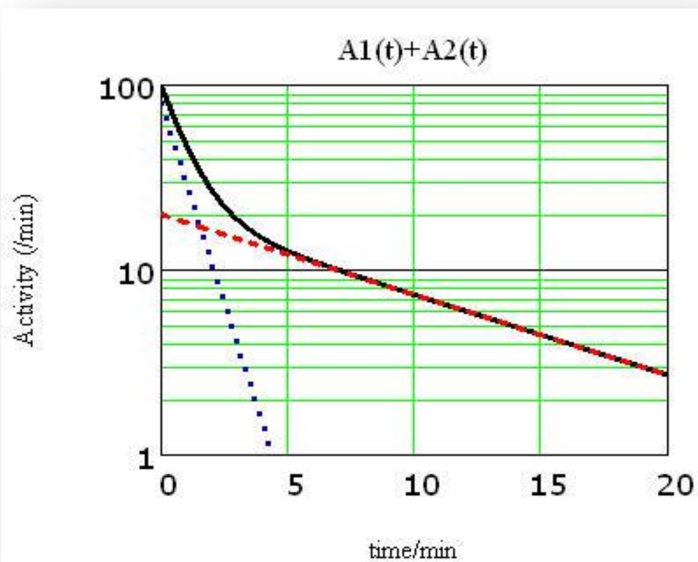
Sample with 2 components (N_1, N_2) → same type of radiation (γ-rays)



$$A_i(t) = A_i(0) \cdot e^{-\lambda_i \cdot t} \qquad (i = 1,2)$$

Total activity :

$$A(t) = A_1(0) \cdot e^{-\lambda_1 \cdot t} + A_2(0) \cdot e^{-\lambda_2 \cdot t}$$

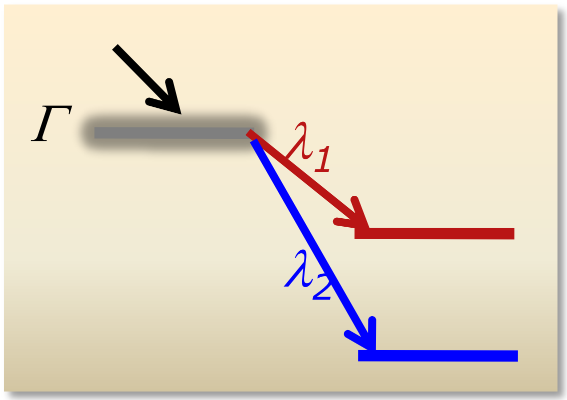


Decompose total decay curve → λ_1, λ_2.

Simultaneous fit or deduce constant λ_2 for "shallow" decay first

Branching Decay

Genetically dependent species: Sample depopulated by 2 decay paths (λ_1, λ_2)



$$\lambda = \lambda_1 + \lambda_2$$

$$\Gamma = \Gamma_1 + \Gamma_2 \quad \text{"level width"}$$

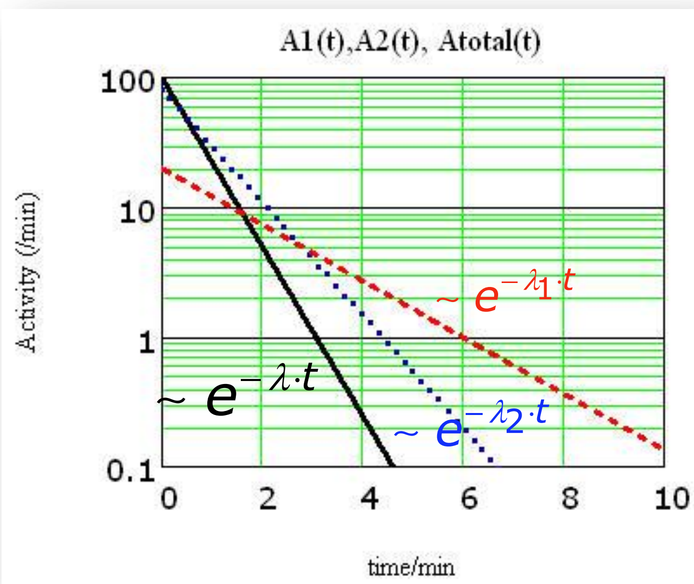
$$\frac{dN(t)}{dt} = -\lambda \cdot N(t) = -(\lambda_1 + \lambda_2) \cdot N(t)$$

$$N(t) = N(0) \cdot e^{-\lambda \cdot t} = N(0) \cdot e^{-(\lambda_1 + \lambda_2) \cdot t}$$

$$A(t) = \lambda \cdot N(t) = \lambda \cdot N(0) \cdot e^{-\lambda \cdot t} = A_1(t) + A_2(t)$$

Partial activities:

$$\rightarrow A_i(t) = \lambda_i \cdot N(0) \cdot e^{-\lambda \cdot t} \qquad (i = 1, 2)$$



Partial decay rates/half lives:

$$\frac{A_i(t)}{A(t)} = \frac{\lambda_i \cdot N(t)}{\lambda \cdot N(t)} = \frac{\lambda_i}{\lambda} \qquad \left(t_{1/2}\right)_i = \frac{0.693}{\lambda_i}$$

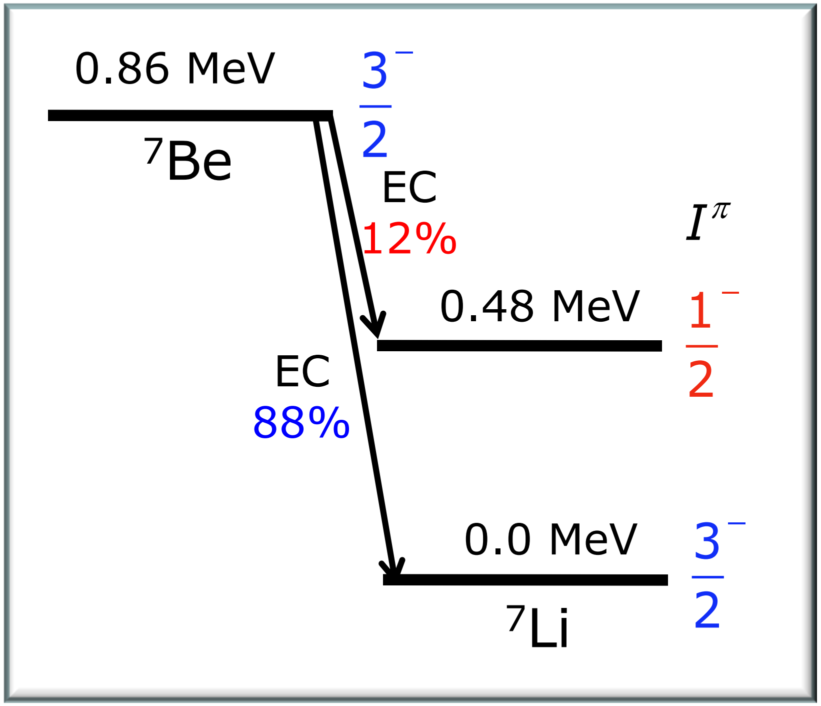
Identify radiation type *i* to measure partial decay rates/half lives.

Branching in EC β Decay

ν phase space depends on
$Q = E_{max}$ →
rate λ increases with E_{max}

$$P_{if} = G_F^2 \frac{2 \cdot Z^3}{\pi^2 \hbar^4 c^3 a_B^3} E_\nu^2 \qquad E_\nu = E_{max} = Q$$

$$\lambda\left(E_{max}\right) \propto E_{max}^2$$



$$\frac{\lambda_{ex}\left(0.478\,MeV\right)}{\lambda_{gs}} = \frac{\left(Q - 0.478MeV\right)^2}{Q^2}$$

$$\frac{\lambda_{ex}}{\lambda_{gs}} = \left(\frac{0.382}{0.861}\right)^2 = 0.20$$

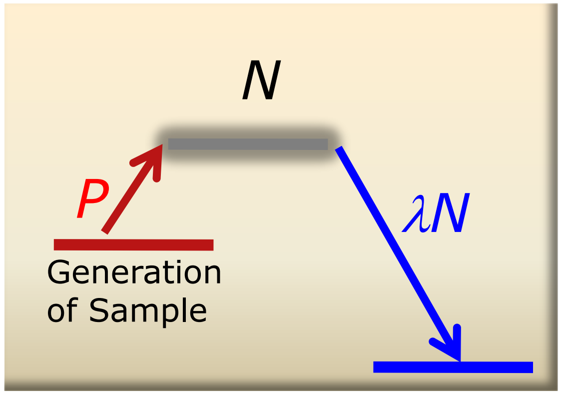
Experimental value correct order of magnitude but disagrees quantitatively

$$\left(\frac{\lambda_{ex}}{\lambda_{gs}}\right)_{exp} = 0.115$$

Reason: $\psi_n \neq \psi_p$ because of nuclear spin change $3^-/2 \rightarrow 1^-/2$ weaker magnetic transition

Activation and Decay

Competition production/decay for a species with *N(t) members,*
Example of genetically related decay chain.



Irradiation of sample produces unstable species *N.*

Constant rate of production *P=const.*

Constant decay rate λ

Gain- Loss Equation

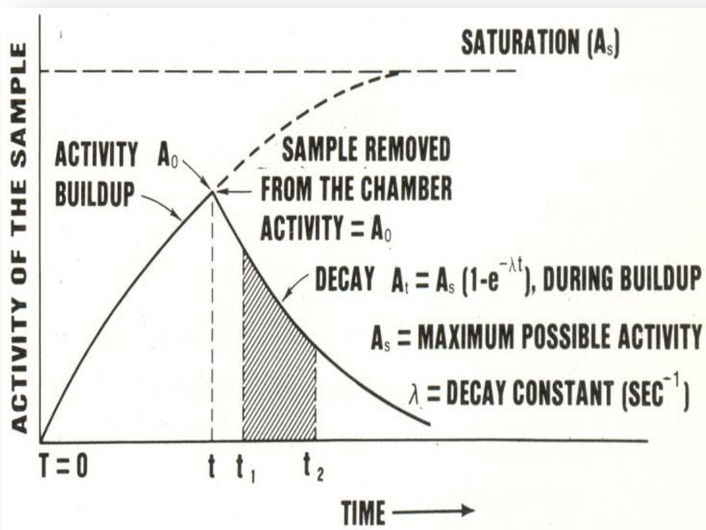
$$\frac{dN(t)}{dt} = -\lambda N(t) + P$$

$$N(t) = \frac{P}{\lambda}\left(1 - e^{-\lambda \cdot t}\right) \rightarrow$$

$$A(t) = P\left(1 - e^{-\lambda \cdot t}\right)$$

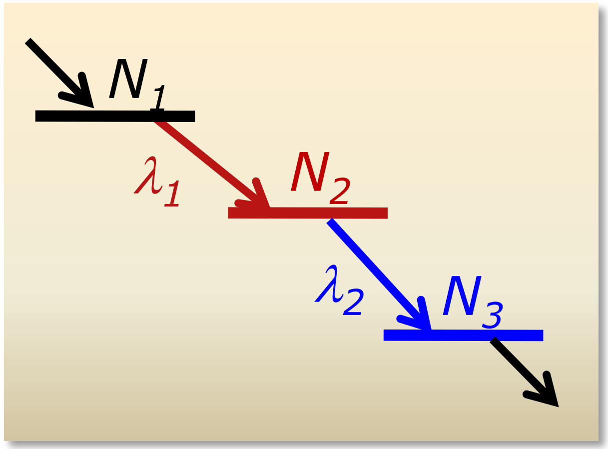
For long times, $t \rightarrow \infty$

$$A(t) = P\left(1 - e^{-\lambda \cdot t}\right) \rightarrow P$$



Generation inefficient for $t \gtrsim 3\ \tau$

Genetically Related Decay Chain



$$\frac{dN_i(t)}{dt} = \lambda_{i-1}N_{i-1}(t) - \lambda_i N_i(t)$$

Gain and loss for *i-th* daughter

Coupled DEq. For populations N_i of nuclei in chain

$$N_1(t) = c_{11} \cdot e^{-\lambda_1 \cdot t} \qquad P(parent)$$

$$N_2(t) = c_{21} \cdot e^{-\lambda_1 \cdot t} + c_{22} \cdot e^{-\lambda_2 \cdot t} \quad P(1.daughter)$$

$$\vdots$$

$$N_k(t) = \sum_{m=1}^{k} c_{km} \cdot e^{-\lambda_m \cdot t} \qquad P((k-1).daughter)$$

$k+1:$ *final grand daughter*

Boundary condition

$$N_i(0) = c_{i1} + c_{i2} + \ldots + c_{ii}$$

$\rightarrow$ *determines* c_{ii}

Recursion Relations

$$c_{ij} = c_{i-1,j} \cdot \frac{\lambda_{i-1}}{\lambda_i - \lambda_j}$$

$$k = 1: \quad N_1(t) = N_1(0) \cdot e^{-\lambda_1 \cdot t}$$

$$N_2(t) = N_1(0)\frac{\lambda_1}{\lambda_2 - \lambda_1} \cdot \left(e^{-\lambda_1 \cdot t} - e^{-\lambda_2 \cdot t}\right)$$

Check by differentiation

Activities and Equilibrium in Decay Chains

$$k = 2: \quad N_1(t) = N_1(0) \cdot e^{-\lambda_1 \cdot t}$$

$$N_2(t) = N_1(0) \frac{\lambda_1}{\lambda_2 - \lambda_1} \cdot \left(e^{-\lambda_1 \cdot t} - e^{-\lambda_2 \cdot t}\right)$$

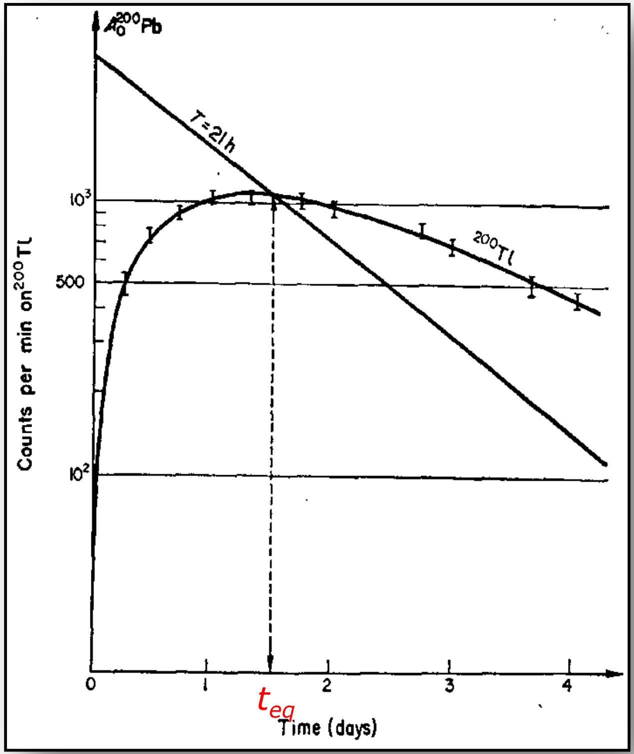
$$N_3(t) = N_1(0) \left\{1 + \frac{\lambda_1}{\lambda_2 - \lambda_1} e^{-\lambda_2 \cdot t} - \frac{\lambda_2}{\lambda_2 - \lambda_1} e^{-\lambda_1 \cdot t}\right\}$$

$$A_1(t) = \lambda_1 N_1(t) = A_1(0) \cdot e^{-\lambda_1 \cdot t} = -\frac{dN_1}{dt}$$

$$A_2(t) = \lambda_2 N_2(t) = A_1(0) \frac{\lambda_2}{\lambda_2 - \lambda_1} \cdot \left(e^{-\lambda_1 \cdot t} - e^{-\lambda_2 \cdot t}\right)$$

$$A_2(t) \neq -\frac{dN_2}{dt} \qquad A_3(t) = 0 \quad (\lambda_3 = \infty)$$

$$\frac{A_2(t)}{A_1(t)} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \cdot \left(1 - e^{-(\lambda_2 - \lambda_1) \cdot t}\right) \xrightarrow[t \to \infty]{} \left(\frac{\lambda_2}{\lambda_2 - \lambda_1}\right)$$



Transitory/secular Equilibrium

$$A_1(t_{eq}) = A_2(t_{eq}) \quad \rightarrow \quad t_{eq} = \frac{\ell n(\lambda_1/\lambda_2)}{(\lambda_1 - \lambda_2)}$$

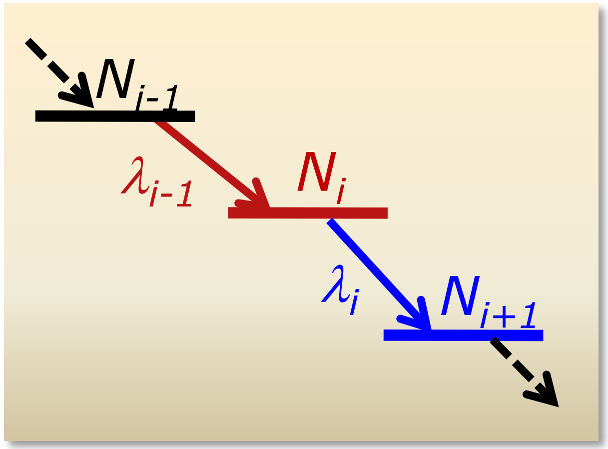
^{200}Pb: $t_{1/2}$=21h → ^{200}Tl: $t_{1/2}$=26h → ^{200}Hg

$$\lambda_1 = 0.693/21h = 9.17 \cdot 10^{-6}\, s^{-1} > \lambda_2$$

$$\lambda_2 = 0.693/26.4h = 7.29 \cdot 10^{-6}\, s^{-1}$$

$$t_{eq} = \frac{0.229}{1.88 \cdot 10^{-6} s^{-1}} = 1.22 \cdot 10^5 s = 1.41d$$

Secular Equilibrium in a Decay Chain



$$\frac{dN_i(t)}{dt} = \lambda_{i-1}N_{i-1}(t) - \lambda_i N_i(t)$$

Gain and loss for *i-th* daughter

Population N_i of daughter *i* in chain

$$N_i(t) = c_1 \cdot e^{-\lambda_1 \cdot t} + c_2 \cdot e^{-\lambda_2 \cdot t} + ... + c_i \cdot e^{-\lambda_i \cdot t}$$

$$c_1 = \frac{\lambda_1 \cdot \lambda_2 \cdot \cdots \cdot \lambda_{i-1}}{(\lambda_2 - \lambda_1) \cdot \cdots \cdot (\lambda_i - \lambda_1)} N_1(0), \quad c_2 = ...$$

Chain survives for long time, if $\lambda_1 \ll \lambda_i$, for all $i > 2$. *Only term* $\sim e^{-\lambda_1 \cdot t}$ *survives.*

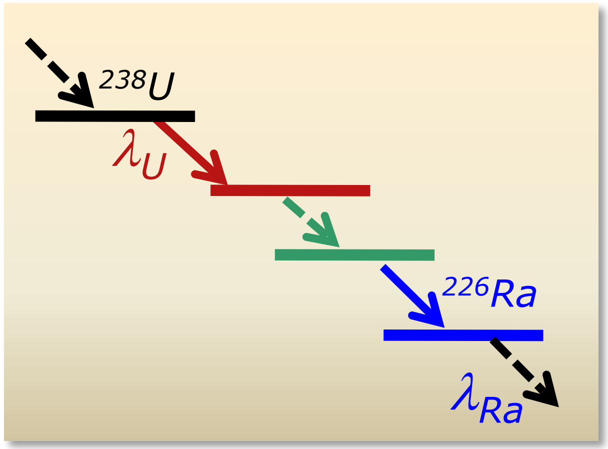
$$N_i(t) \approx c_1 \cdot e^{-\lambda_1 \cdot t} \quad with\ c_1 = \frac{\lambda_1 \cdot \lambda_2 \cdot \cdots \cdot \lambda_{i-1}}{(\lambda_2 - \lambda_1) \cdot \cdots \cdot (\lambda_i - \lambda_1)} N_1(0)$$

$$\frac{A_i(t)}{A_1(t)} = \underbrace{\frac{\lambda_2}{(\lambda_2 - \lambda_1)}}_{\approx \lambda_2} \cdot \cdots \cdot \underbrace{\frac{\lambda_i}{(\lambda_i - \lambda_1)}}_{\approx \lambda_i} \rightarrow \frac{A_i(t)}{A_1(t)} = \prod_{j=2}^{i} \frac{\lambda_j}{(\lambda_j - \lambda_1)} \approx 1$$

Secular Equilibrium

$$\lambda_1 N_2(t) \approx \lambda_2 N_2(t) \approx .... \approx \lambda_i N_i(t)$$

Example: Determination of ^{238}U Lifetime



Extremely long lifetime of ^{238}U → direct measurement difficult

One of the decay products is

^{226}Ra with $t_{1/2}$ = 1620 *a*

Relative abundance $N_U/N_{Ra} = 2.8 \cdot 10^6$

Secular Equilibrium

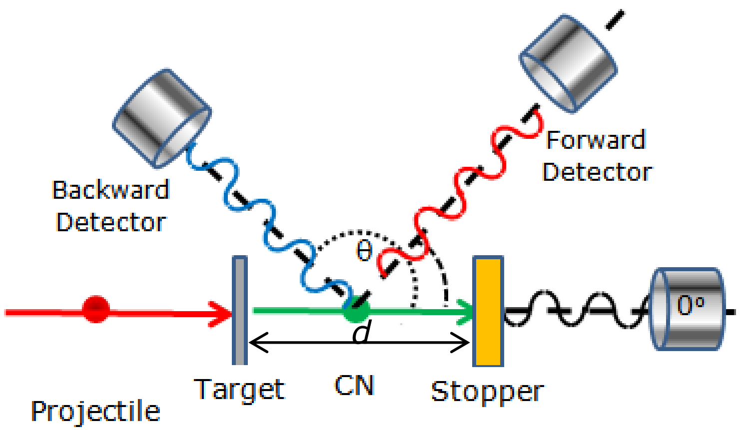
$$\lambda_1 N_2(t) \approx \lambda_2 N_2(t) \approx .... \approx \lambda_i N_i(t) \quad \rightarrow \quad \lambda_U N_U(now) \approx \lambda_{Ra} N_{Ra}(now)$$

$$\lambda_U \approx \frac{N_{Ra}(now)}{N_U(now)} \lambda_{Ra} \quad \rightarrow \quad \tau_U \approx \frac{N_U(now)}{N_{Ra}(now)} \tau_{Ra} \qquad t_{1/2}(U) \approx \frac{N_U(now)}{N_{Ra}(now)} t_{1/2}(Ra)$$

$$t_{1/2}\left(^{238}U\right) \approx 2.8 \cdot 10^6 \cdot 1620\, a = 4.5 \cdot 10^9\, a$$

Recoil Distance Doppler Shift Method

Mean Lifetime Determination of the ^{106}Cd $I^{\pi} = 2^{+}_{1}$ state,



Doppler Shift $$E_{\gamma}(\theta) = E_{\gamma}^{Stop}\left[1 + \frac{\upsilon_{CN}}{c}\cdot\cos\theta\right]$$

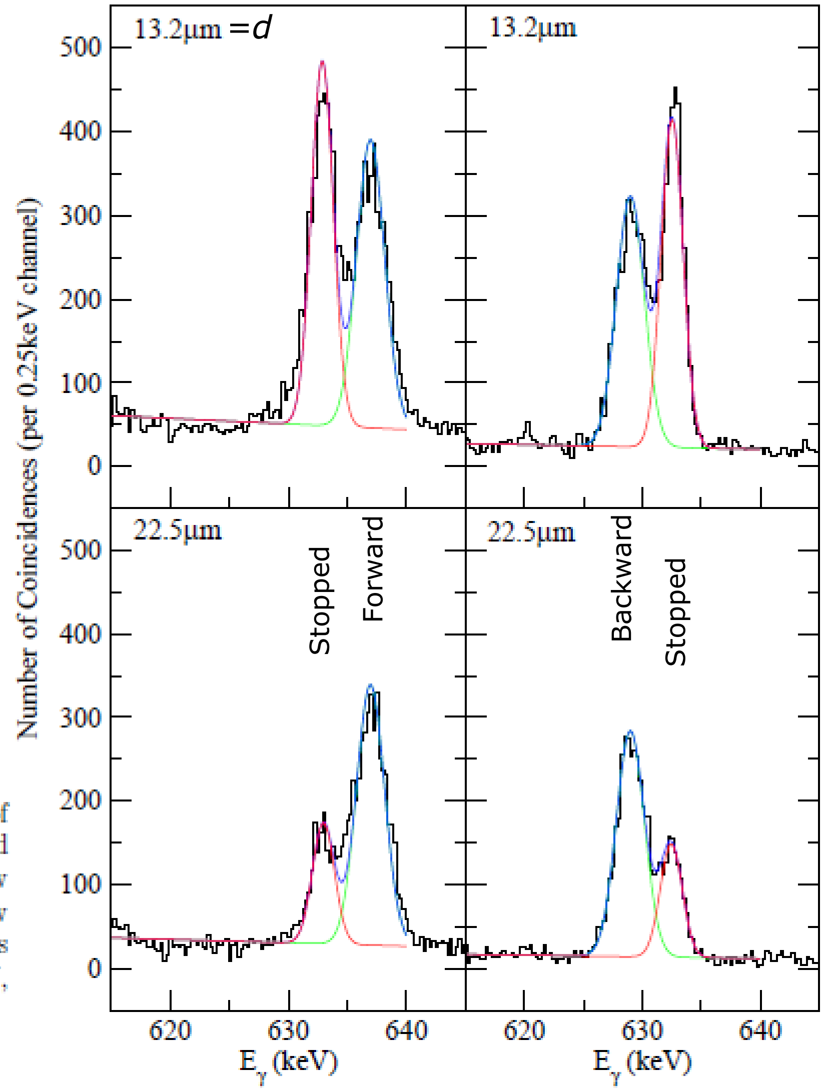
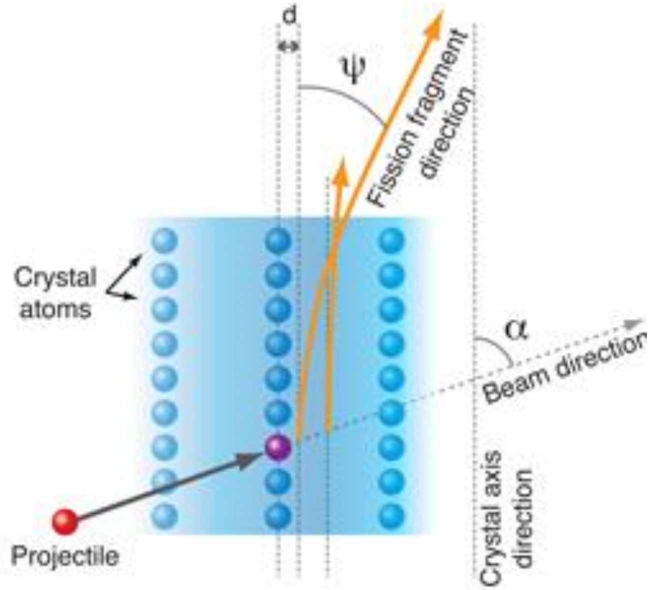
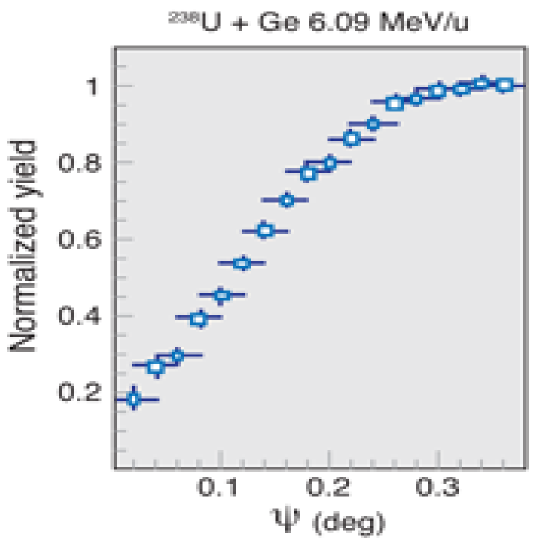


Figure 4.36: Left Hand Side Spectra: Stopped and forward-shifted components of the 633 keV, $I^{\pi} = 2^{+}_{1} \rightarrow 0^{+}_{1}$, transition. Right Hand Side Spectra: Stopped and backward-shifted components of the 633 keV, $I^{\pi} = 2^{+}_{1} \rightarrow 0^{+}_{1}$, transition. Top Row Spectra: Projections taken at a target-stopper distance of 13.2 μm. Bottom Row Spectra: Projections taken at a target-stopper distance of 22.5 μm. All projections have been generated from a gate on the backward-shifted component of the 861 keV, $I^{\pi} = 4^{+}_{1} \rightarrow 2^{+}_{1}$, transition.

Crystal Blocking Technique





Principle of the crystal blocking technique.

Heavy ions bombard a single-crystal target, form CN.
CN fissions with lifetime ~10^{-18} s.
FF emitted in the plane of the target atoms (ψ=0) are blocked from reaching the detector.
FF emitted from recoiling nuclei that survive long enough to move into a channel between the crystal planes (distance d) are detected with little energy loss.

Thermal vibrations in the crystal determine the lower time limit for blocking.

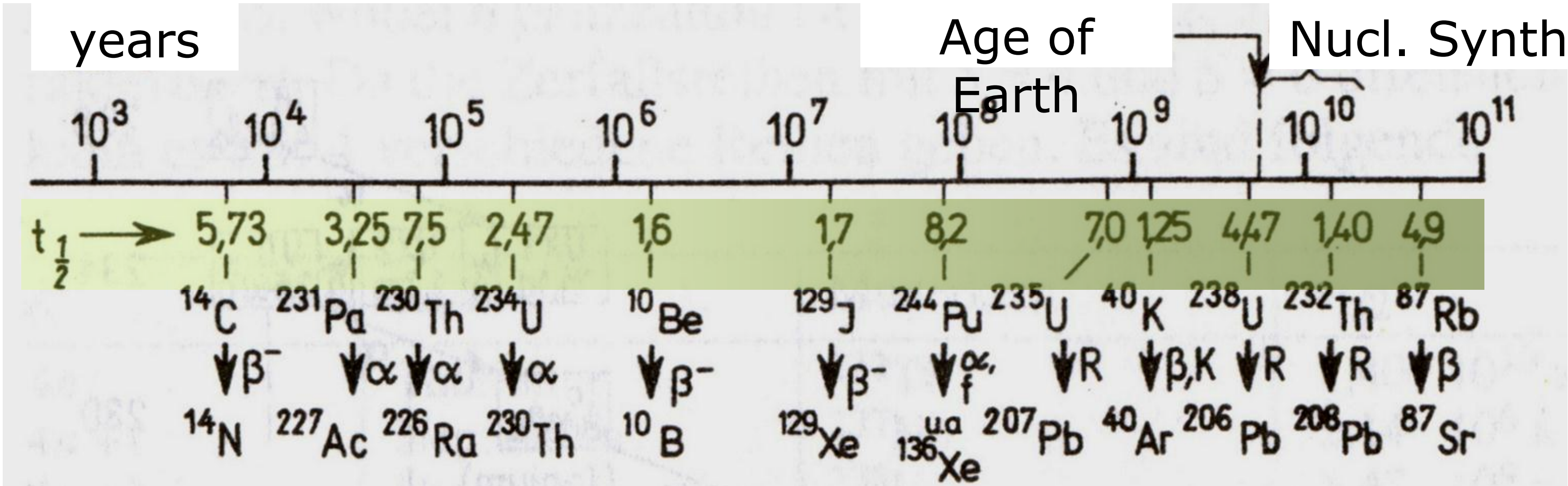
^{238}U+Ge @ E/A= 6.09 MeV

Blocking dip observed for Z=124,
FF 67<Z_{FF}<85.
The width of the dip depends on atomic number and kinetic energy of the fission fragment

M. Morjean et al., Phys. Rev. Lett. **101**, 072701 (2008)

Age Determination
(Dating)

Halflives of Radio-Isotopes for Dating

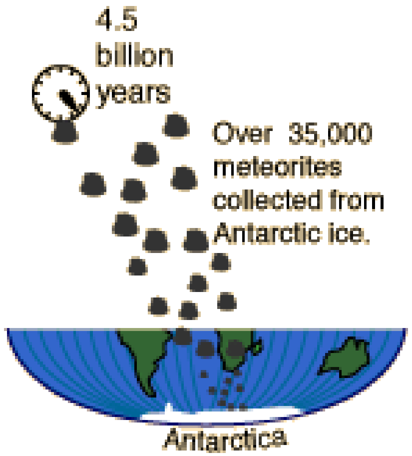


α, β, β^- : particles measured to identify fractional abundance of radioactive isotope,

K: K electron capture

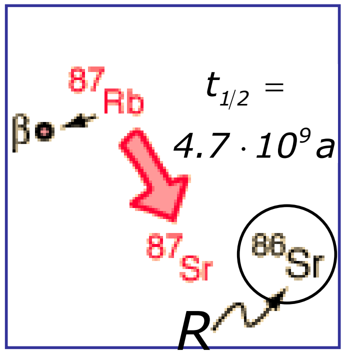
R : measure series of several decay products

Rb/Sr Dating of Rocks/Age of the Earth



Undisturbed by erosion/transmutation

All rocky objects (planets, asteroids, meteorites) of solar system crystallized ≈ simultaneously (*t=0*) out of interstellar dust/nebula (supernova remnants).



$$Parent\ P = {}^{87}Rb,\ daughter\ D = {}^{87}Sr$$

$$Reference\ R = {}^{87}Rb\quad (stable)$$

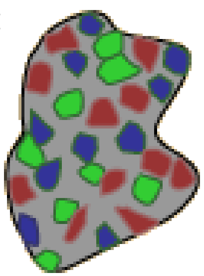
$$N_P(0) = N_P(t) + N_D(t) \qquad \text{but unknown!}$$

$$N_P(t) = N_P(0)\cdot e^{-\lambda\cdot t} \qquad N_R(t) = N_R(0)$$

$$N_D(t) = N_P(0) - N_P(t) + N_D(0)$$

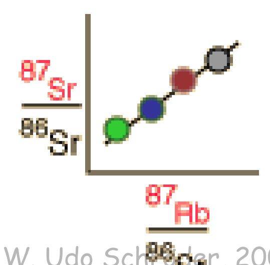
$$N_D(t) = N_P(t)\cdot\left[e^{+\lambda\cdot t} - 1\right] + N_D(0)$$

$$\underbrace{\frac{N_D(t)}{N_R(t)}}_{y} = \underbrace{\frac{N_P(t)}{N_R(t)}}_{x}\cdot\underbrace{\left[e^{+\lambda\cdot t} - 1\right]}_{m} + \underbrace{\frac{N_D(0)}{N_R(0)}}_{y_0}$$



Different minerals in meteorite containing different amounts of N_P → different x

Construct isochron

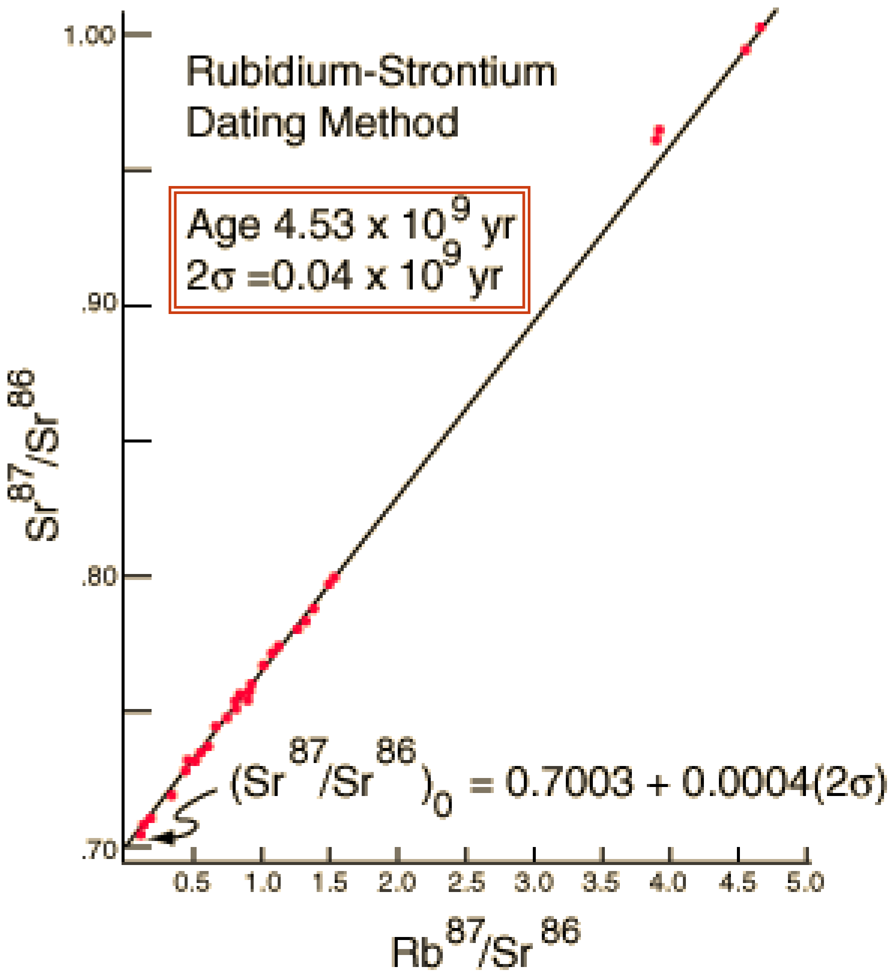


$$y = y_0 + m(t)\cdot x$$

$$\rightarrow t = \frac{1}{\lambda}\cdot \ell n\left[m + 1\right]$$

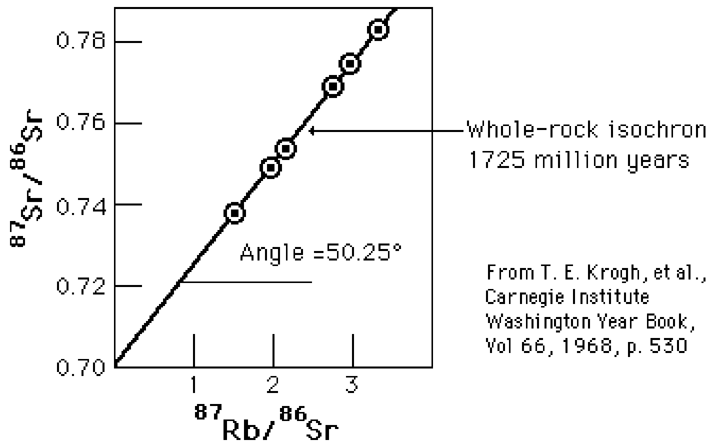
Age of rock (since formation)

Age of the Earth



G. W. Wetherill, Ann. Rev. Nucl. Sci. 25, 283 (1975)

Age of Earth = $4.5 \cdot 10^9$ *a*
Moon has similar age

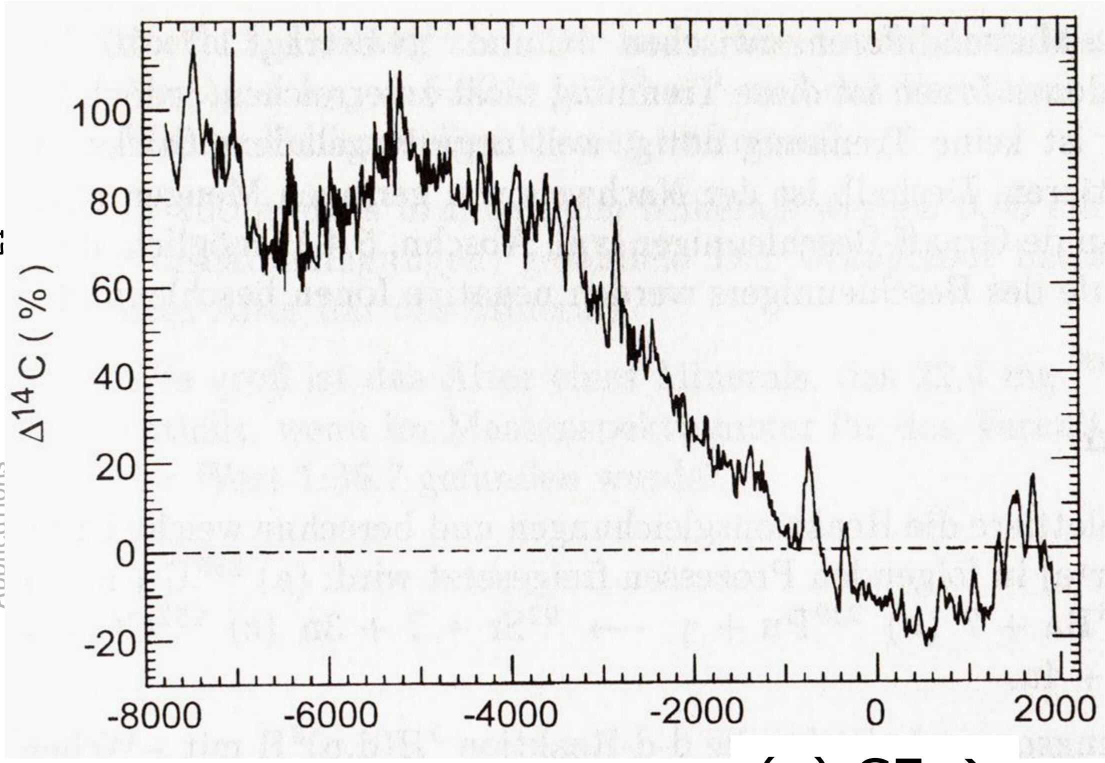


Whole-rock rubidium-strontium isochron for a set of samples of a Precambrian granite body exposed near Sudbury, Ontario.

Terrestrial volcanic activity dated: produces younger rocks

Calibration of ^{14}C Dating Methods

Variation in ^{14}C Production



(a) CE →

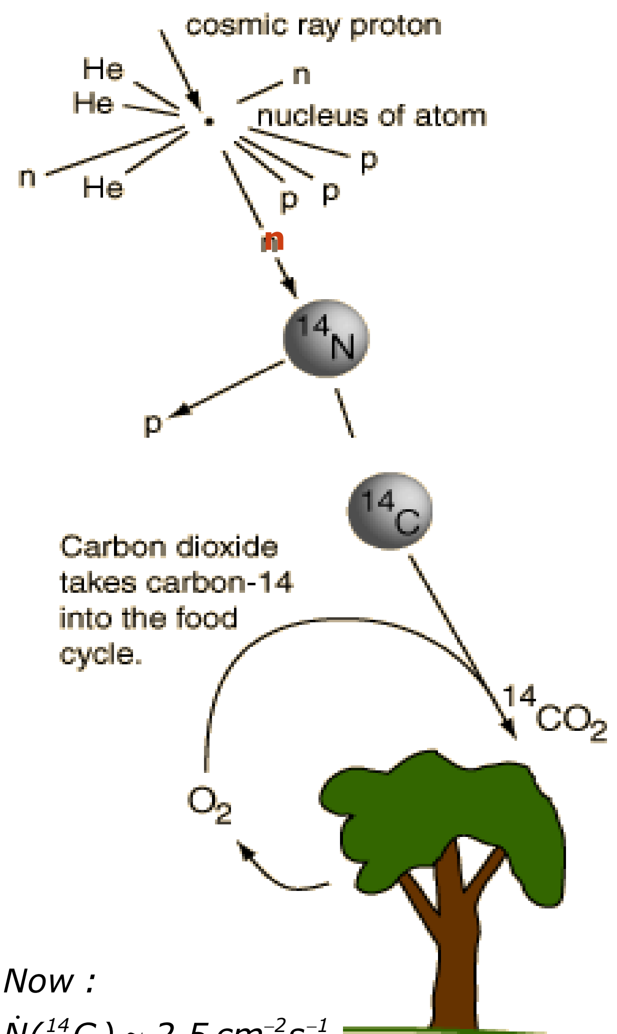
t-dependent flux of cosmic rays (solar cycles)
→ *t*-dependent ^{14}C production and intake

Calibration:

^{14}C-analyze yearly rings in trees of different ages (number and widths of rings), connect to fossils

Errors in very old samples lead to underestimation of age (few hundred years).

Carbon Dating of Organic Objects



Now :

$\dot{N}(^{14}C) \approx 2.5\,cm^{-2}s^{-1}$

$^{14}C/^{12}C = 1.5 \cdot 10^{-12}$

$t_{1/2} = 5730\,a$

$$\lambda = \frac{0.6931}{t_{1/2}} = \frac{0.6931}{5730a} = 1.21 \times 10^{-4}\,a^{-1}$$

$$N_{12C}(t) = N_{12C}(t=0)$$

$$N_{14C}(t) = N_{14C}(t=0) \cdot e^{-\lambda \cdot t}$$

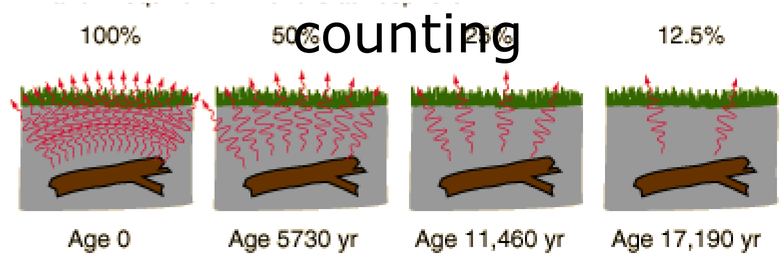
$$R(t) = \frac{N_{14C}(t)}{N_{12C}(t)} = \underbrace{R(t=0)}_{\approx 1.3 \cdot 10^{-12}} \cdot e^{-\lambda \cdot t}$$

$t = 0$:
time of death
No further
^{14}C intake

$$\rightarrow \boxed{"age" = t = \frac{1}{\lambda}\,\ell n\left[\frac{R(0)}{R(t)}\right]}$$

Measure $^{14}C/^{12}C$ ratio of sample at *t*

Conventional method: β counting



Direct ^{14}C counting method: Accelerator Mass Spectroscopy → $R \gtrsim 10^{-16} \quad (10^{5}\,a)$